

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In re Patent Application of:

T. YAMAGUCHI, et al : Group Art Unit: 1745

Application No. 09/936,148 : Examiner: Laura S. WEINER

Filed: September 7, 2001 :

For: ELECTROLYTE MEMBRANE FOR FUEL CELL AND MANUFACTURING
METHOD THEREOF

DECLARATION UNDER 37 C.F.R. §1.132

Honorable Commissioner of Patents and Trademarks

Washington, D.C. 20231

Sir:

I, Takeo YAMAGUCHI, do declare and state as follows:

I graduated from The University of Tokyo with a Doctor's
Degree of Chemical Engineering in March 1993;

I became a post doctoral research fellow at department
of Chemical Engineering, University of Colorado at Boulder
in April, 1993, and since that time I have been engaged in
research and development in the field of membrane science and
technology and polymer electrolyte fuel cells at Department
of Chemical System Engineering, University of Tokyo;

I am a co-inventor of the subject matter disclosed and

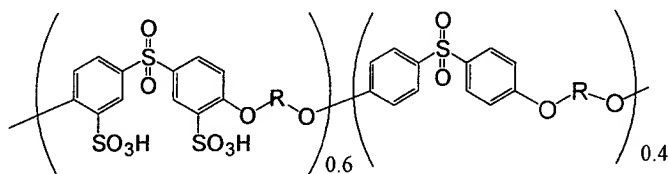
claimed in the above-identified application; and

I am familiar with the Office Action of August 23, 2005, and understand the Examiner's rejections.

The following additional comparative experiment (Comparative Experiment A) was carried out by me or under my supervision, in order to make the differences between the subject matter and cited references more clear and to make advantages of the subject matter more clear.

Comparative Experiment A

SPES polymer represented by following formula was prepared. Furthermore, water is good solvent for SPES polymer having 60 mol% sulfonated unit content.



The prepared SPES polymer powder was resolved in N-methylpyrrolidone. The resulting solution was applied onto a porous polyimide substrate (manufactured by UBE corp., thickness: 30 μ m, porosity: 50%), to obtain a membrane in which SPES polymer was filled in pores of the substrate. The porous polyimide substrate was opaque, while the membrane was transparent, showing that SPES polymer was filled in pores

of the substrate. The filling ratio of SPES polymer in membrane was 60 %, determined by the following calculation equation ($W_{\text{mem}}^{\text{d}}$: weight of membrane; $W_{\text{sub}}^{\text{d}}$: weight of substrate).

$$\text{Filling ratio} = (W_{\text{mem}}^{\text{d}} - W_{\text{sub}}^{\text{d}}) * 100 / W_{\text{sub}}^{\text{d}}$$

The resulting membrane (3 cm * 3 cm) was immersed in hot water (80 °C) for 24 hours. As described above, water is good solvent for SPES polymer (60 mol% sulfonated as illustrated above). Then, change of appearances between before- and after-immerse membrane was observed, and change of weights between before- and after- immerse membrane was determined by the following equation ($W_{\text{mem, after}}^{\text{d}}$: weight of after-immerse membrane; $W_{\text{mem, before}}^{\text{d}}$: weight of before-immerse membrane; and $W_{\text{sub}}^{\text{d}}$: weight of substrate).

Remaining ratio =

$$(W_{\text{mem, after}}^{\text{d}} - W_{\text{sub}}^{\text{d}}) * 100 / (W_{\text{mem, before}}^{\text{d}} - W_{\text{sub}}^{\text{d}})$$

The remaining ratio of the membrane of interesting was 5 % or less, and the appearance of after-immerse membrane was opaque. These results show that polymers in pores of the membrane prepared by the applying method were leached out.

Experiment B

As described in Example 1 of the present application, an electrolyte membrane comprising PTFE and poly(acrylic acid) wherein one end of molecule of poly(acrylic acid) is bound to surface of pores of the electrolyte membrane was prepared. Furthermore, mixed solvent (water/methanol = 10/90 percent by weight) is good solvent for poly(acrylic acid).

The electrolyte membrane was subjected to vapor (water/methanol = 10/90 percent by weight) permeation test at temperatures of 25, 80, 120, 150 and 180 °C. The test at each temperature took about 5 hours (total test time = about 25 hours). The appearance of the electrolyte membrane after the tests did not change. More, the proton-conductivities were not changed between before- and after- test. These results show that poly(acrylic acid) filled in pores of the electrolyte membrane were not leached out since one end of molecule of poly(acrylic acid) is bound to surface of pores of the electrolyte membrane.

Conclusions

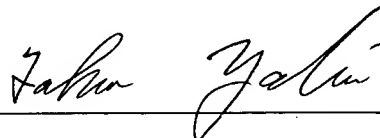
Polymers according to the applying method as described in the above comparative experiment A, Chowdhury et al. (USP No. 5,798,180) or Formato et al. (USP No. 6,248,469) are easily leached out when the membrane comprising the polymers in pores thereof is immersed in good solvent for the polymers, since one end of each of the polymers is not bound to surface of

pores of the membrane. On the other hand, polymers according to the present invention were not leached out, since one end of each of the polymers is bound to surface of pores of the membrane.

In brief, Chowdhury et al. (USP No. 5,798,180) or Formato et al. (USP No. 6,248,469) does not disclose or suggest one of features of the present invention, i.e., "one end of a molecule of polymer is bound to a surface of the pore".

I further declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true, and further, that these statements were made with the knowledge that willful false statements and like so made are punishable by fine or imprisonment, or both, under section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issued thereon.

DATE: Dec. 13, 2005



Takeo Yamaguchi